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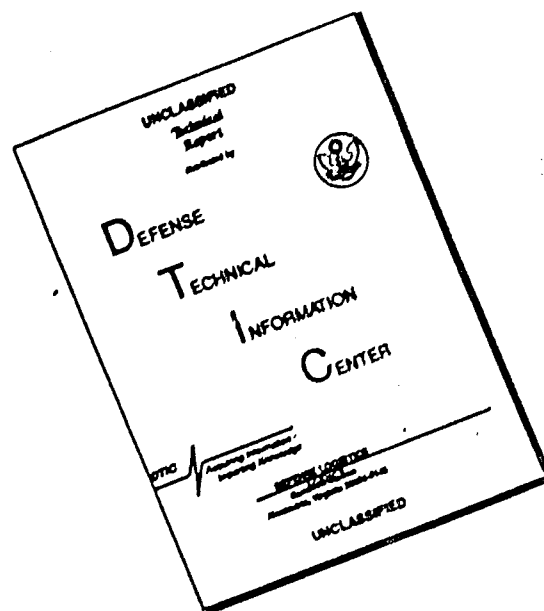
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Technical Report No 1

by

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THE EFFECT OF REACTANT CONSUMPTION IN
NONSTEADY EXPLOSION THEORY

by

William Squire

Technical Report No. 1
Contract No. CST-362
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Prepared for

The National Bureau of Standards
Washington 25, D. C.

Attn: Dr. A. F. Robertson
Chief, Fire Protection Section
Building Research Division

August 1961

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APPROVED:



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SUMMARY

The effect of reactant consumption in nonsteady explosion theory is investigated by an exact numerical method and an approximate analytical treatment. Results are presented for the critical conditions and the ignition delay for supercritical conditions. The approximate analysis gives ignition delays in good agreement with the numerical solution, but the critical conditions are not obtained as accurately as in an alternative analysis due to Thomas. A comparison of temperature-time histories computed by the exact numerical method with experiments reported by Akita showed satisfactory agreement and indicated that reactant consumption was significant for wood.

LIST OF SYMBOLS

ρ	-	density
c	-	specific heat
T	-	temperature
t	-	time
K	-	thermal conductivity
Q	-	heat of reaction per gram of material reacted
w	-	concentration of reactant
k	-	prefactor in Arrhenius rate expression
n	-	order of reaction
E	-	activation energy
R	-	gas constant
θ	-	dimensionless temperature, $\frac{E}{RT_{in}^2} (T - T_{in})$
τ	-	dimensionless time, $Kt/(\text{characteristic length})^2$
δ	-	dimensionless reaction rate, $\frac{w_{in}^{n+1} Q_{ke}^{-E/RT_{in}}}{KRT_{in}^2} \times (\text{characteristic length})^2$
λ	-	dimensionless concentration, w/w_{in}
c	-	RT_{in}/E

LIST OF SYMBOLS (Cont'd)

A - dimensionless heat transfer coefficient

B - dimensionless adiabatic temperature rise, $\frac{E}{RT_{in}^2} \frac{w_{in} Q}{\rho c}$

q - an adjustable parameter

P - a constant in Equation (14)

F() - a function used in the numerical solution defined in Equation (7b)

$\Delta\theta$ - step size in the numerical solution

Subscripts

in - initial

cr - critical

INTRODUCTION

The basic partial differential equations for a homogeneous combustion process with an Arrhenius type reaction rate term are

$$\rho c \frac{\partial T}{\partial t} = K \nabla^2 T - Q \frac{\partial w}{\partial t} \quad (1a)$$

$$\frac{\partial w}{\partial t} = -k w^n e^{-E/RT} \quad (1b)$$

The nonsteady theory is an approximate formulation which replaces the partial differential equations by ordinary differential equations giving the "average" temperature and composition as functions of time. The space variation is eliminated by replacing the Laplacian, which represents the heat conduction, by an equivalent heat loss term proportional to the difference between the "average" temperature and the ambient temperature.

The physical significance of the average quantities and the relation of the heat loss coefficient in the nonsteady theory to the physical boundary condition on (1) has been discussed in recent papers by Gray and Harper¹ and by Thomas²

In terms of dimensionless variables (defined in the List of Symbols) the ordinary differential equations are

$$\frac{d\theta}{d\tau} = \delta \lambda^n e^{\frac{\theta}{1+c\theta}} - A\theta \quad (2a)$$

and

$$\frac{d\lambda}{d\tau} = -\frac{\delta}{B} \lambda^n e^{\frac{\theta}{1+\epsilon\theta}} \quad (2b)$$

subject to the conditions $\theta = 0$ and $\lambda = 1$ at $\tau = 0$. The form of the equation is independent of geometry which only enters in the determination of the heat transfer coefficient, A .

The object of this paper is to present approximate analytical and numerical solutions of (2). While there have been a number of previous investigations, simplifications have been made, especially of (2). While these approximations are justified for gaseous reactions, which were the motivation for the earlier work, Thomas³ has pointed out that for solids, such as wood, where B is smaller by an order of magnitude, it is important to take the reactant consumption into account.

It is well known that (2) has two types of solutions depending on the value of A/δ . For large values of A/δ , the heat generated by the reaction can be conducted away, and there is only a small temperature rise, the maximum value of θ being about 1. This type of solution will be referred to as stable or subcritical. For small values of A/δ , unstable or supercritical solutions are obtained. The heat cannot be conducted away rapidly enough, and θ approaches the adiabatic value B . The transition is quite sharp for large values of B . The important problems are:

- (1) To determine the critical value of A/δ dividing the two types of solution, and
- (2) To determine the time at which the rapid temperature rise begins in the supercritical case. Conventionally,

3

this ignition delay is defined as the time at which

$\theta = 2$. For materials such as wood with a small heat

of reaction, there is no explosion in the literal sense

but the phenomenon is similar in principle to the thermal

explosion of gases.

ANALYSIS

Both the approximate analytical solution and the numerical solution take advantage of the absence of τ from (2). Multiplying (2b) by $n\lambda^{n-1}$ and dividing by (2a) gives the differential equation

$$\frac{d\lambda^n}{d\theta} = -\frac{n}{B} \frac{\lambda^{2n-1} e^{\frac{\theta}{1+\epsilon\theta}}}{\lambda^n e^{\frac{\theta}{1+\epsilon\theta}} - \frac{A}{\delta} \theta} \quad (3)$$

subject to $\lambda = 1$ at $\theta = 0$. After solving this equation either analytically or numerically, the solution can be expressed as a function of τ by the auxiliary relation

$$\delta\tau = \int_0^\theta \frac{d\theta}{\lambda^n e^{\frac{\theta}{1+\epsilon\theta}} - \frac{A}{\delta} \theta} \quad (4)$$

The use of θ as the independent variable involves some problems as both λ and τ are double valued functions of θ ; however, these are not significant in the specific problems under consideration, and it was found easier to work with (3) than with the alternate form

$$\frac{d\theta}{d\lambda} = -B \left[1 - \frac{A}{\delta} \frac{\theta}{\lambda^n} e^{-\frac{\theta}{1+\epsilon\theta}} \right] \quad (5)$$

Numerical Solution

In this section, only a first order equation ($n = 1$) is considered, and ϵ is neglected. With these simplifications, (3) can be written as an integral equation

$$\lambda = 1 - B^{-1} \int_0^\theta \frac{\lambda d\theta}{\lambda - \frac{A}{\delta} \theta e^{-\theta}} \quad (6)$$

This integral form is only valid while θ increases to its maximum value. However, no difficulty results from this limitation. In subcritical cases, an imaginary value of λ is obtained when the calculation is carried past the maximum value of θ . In the supercritical cases, the calculation is only carried out to $\theta = 2$ which is well below the maximum value.

The numerical solution is based on the relations

$$2\lambda(\theta) = \frac{\frac{A}{\delta} \theta e^{-\theta} + F(\theta - \Delta\theta) - \frac{\Delta\theta}{2B}}{\left[\left(\frac{A}{\delta} \theta e^{-\theta} + F(\theta - \Delta\theta) - \frac{\Delta\theta}{2B} \right)^2 - 4 \frac{A}{\delta} \theta e^{-\theta} F(\theta - \Delta\theta) \right]^{1/2}} \quad (7a)$$

where

$$F(\theta - \Delta\theta) = \lambda(\theta - \Delta\theta) - \frac{\Delta\theta}{2B} \frac{\lambda(\theta - \Delta\theta)}{\lambda(\theta - \Delta\theta) - \frac{A}{\delta} (\theta - \Delta\theta) e^{-(\theta - \Delta\theta)}} \quad (7b)$$

This procedure is equivalent to the well-known modified Euler method⁴ of solving an ordinary differential equation. The iterative process was eliminated by the assumption $n = 1$ which made the nonlinear algebraic relation a quadratic which was solved to give an explicit formula for $\lambda(\theta)$ in terms of $\lambda(\theta - \Delta\theta)$ and θ . The method is also applicable with ϵ taken into account. The simplifying assumption was made because ϵ is in fact small and the approximate analytical solutions, with which the numerical solution will be compared, neglect ϵ .

It can be seen that the numerical solution of the basic equations only solves the first problem determining the critical value of A/δ for a given value of B , indirectly. The critical value of A/δ for a given value of B can be bounded as closely as desired by means of numerical solutions, but no explicit formula for the critical curve can be obtained. It is, therefore, desirable to consider approximate analytical solutions.

Approximate Analytical Solution

The approximate analytical solution of (6) is based on the method of successive approximations. If an approximate expression for $\lambda(\theta)$ is put into the integral on the right-hand side of (6), an improved approximation is obtained by evaluating the integral. The first approximation will be taken as

$$\lambda = 1 - \frac{q}{B} \theta \quad (8)$$

where q is a parameter to be determined by matching the first and second approximations at a reasonable value of θ . Using the initial slope gives $q = 1$, but the higher value obtained by a matching procedure appears desirable.

It should also be noted that within the accuracy of the approximation

$$\lambda^n \approx 1 - \frac{nq}{B} \quad (9)$$

Therefore, the approximate analysis can be extended to an n th order reaction by replacing B by B/n except when B is small.

Examination shows that with λ given by (8), or even with $\lambda = 1$, the integral in (6) cannot be evaluated in closed form. For $\lambda = 1$ (no reactant consumption) Gray and Harper¹ used the approximation

$$e^\theta \approx 1 + .72\theta + \theta^2 \quad (10)$$

This does not allow integration in closed form when (8) is used, therefore, an analogous approximation

$$e^\theta \approx \frac{1 + .718\theta}{1 - .368\theta} \quad (11)^*$$

*The constants $.718 \approx e - 2$ and $.368 \approx 1/e$ were selected to give the correct result for $B \rightarrow \infty$.

was introduced in order to obtain an integrable form. Fig. 1, which compares the two approximations within the range $0 \leq \theta \leq 2$ shows that they are of comparable accuracy.

With these approximations, the relations

$$\lambda = 1 - B^{-1} \int_0^\theta \frac{1 + \left(.718 - \frac{q}{B} \right) \theta - .718 \frac{q}{B} \theta^2}{1 - \left(\frac{A}{\delta} + \frac{q}{B} - .718 \right) \theta + \left(.368 \frac{A}{\delta} - .718 \frac{q}{B} \right) \theta^2} d\theta \quad (12a)$$

and

$$\delta\tau = \int_0^\theta \frac{1 - .368\theta}{1 - \left(\frac{A}{\delta} + \frac{q}{B} - .718 \right) \theta + \left(.368 \frac{A}{\delta} - .718 \frac{q}{B} \right) \theta^2} d\theta \quad (12b)$$

are obtained. These integrals can be evaluated in closed form by well-known methods, though the resulting expressions are rather long.

The Critical Case

The critical case corresponds to the denominator in (12), being a perfect square. Algebraic manipulations gives the relation

$$\left(\frac{A}{\delta} \right)_{cr} = \left(1.454 - \frac{q}{B} \right) + \sqrt{\left(1.454 - \frac{q}{B} \right)^2 - \left(.718 \frac{q}{B} \right)^2} \quad (13)$$

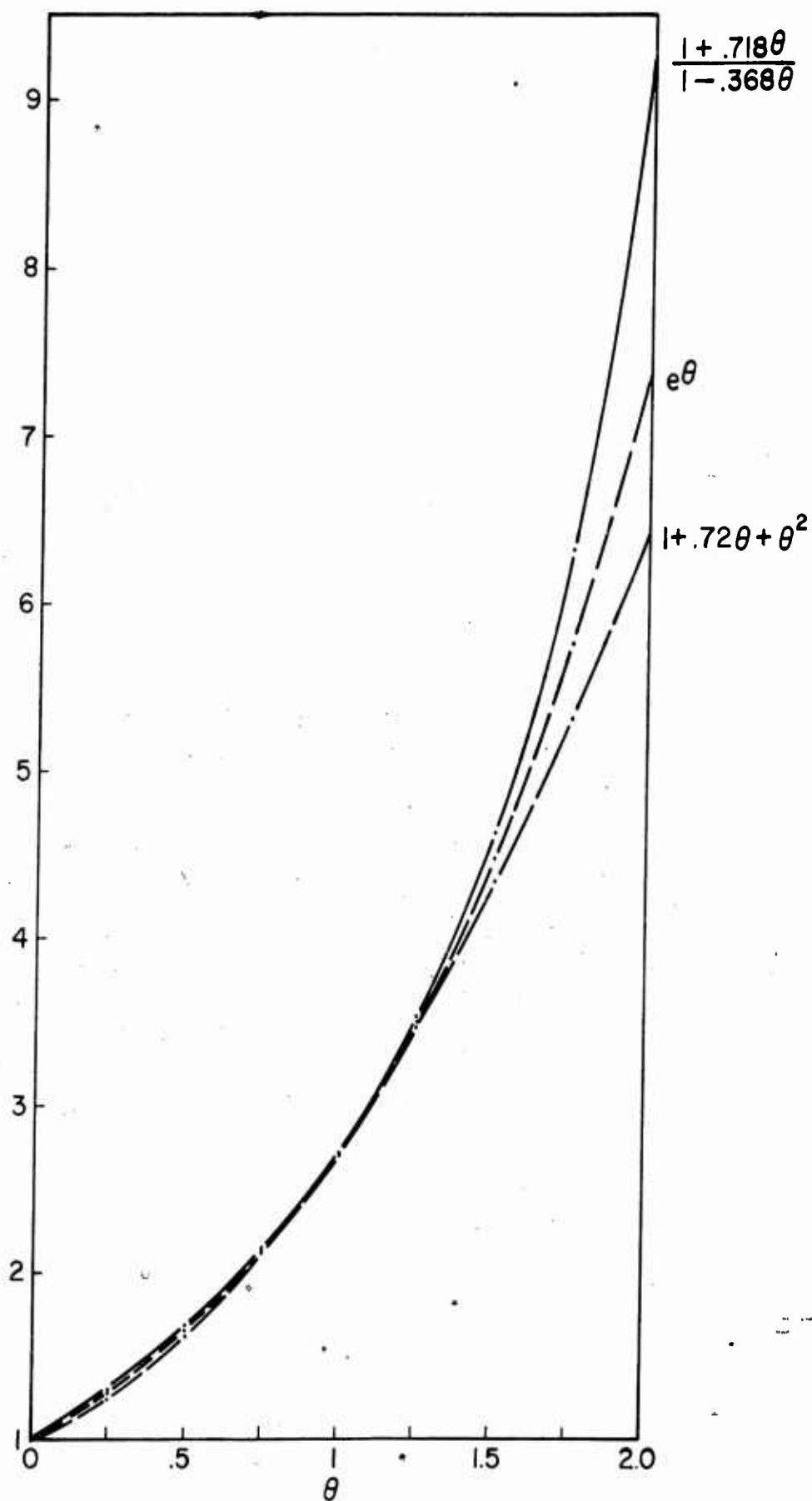


FIGURE 1. COMPARISON OF APPROXIMATIONS TO e^θ

for the critical value of A/δ . However, the expression does not give a definite result until q is evaluated. Introducing (13) for A/δ into (12a) gives

$$\lambda = \int_0^{P\theta} \frac{1 + P^{-1} \left(.718 - \frac{q}{B} \right) (P\theta) - .718 P^{-2} \frac{q}{B} (P\theta)^2}{(1 - P\theta)^2} d(P\theta) \quad (14)$$

where

$$P = \sqrt{.368 \left(\frac{A}{\delta} \right)_{cr} - .718 \frac{q}{B}}$$

Physically, P^{-1} represents the maximum value of θ in the critical case. Therefore, using a constant value of $P\theta$ as the matching point appears plausible. Fig. 2 shows the variation of q with B for several choices of values of $P\theta$ at which (8) and (14) match. It can be seen that q is constant until B becomes relatively small. A discussion of the resulting critical curve will be given below.

Supercritical Case

In the supercritical case, the value of A/δ is arbitrary, provided it is below the critical value. The problem is to compute the value of $\delta\tau$ for $\theta = 2$ by evaluation of (12b). It is reasonable to evaluate q by

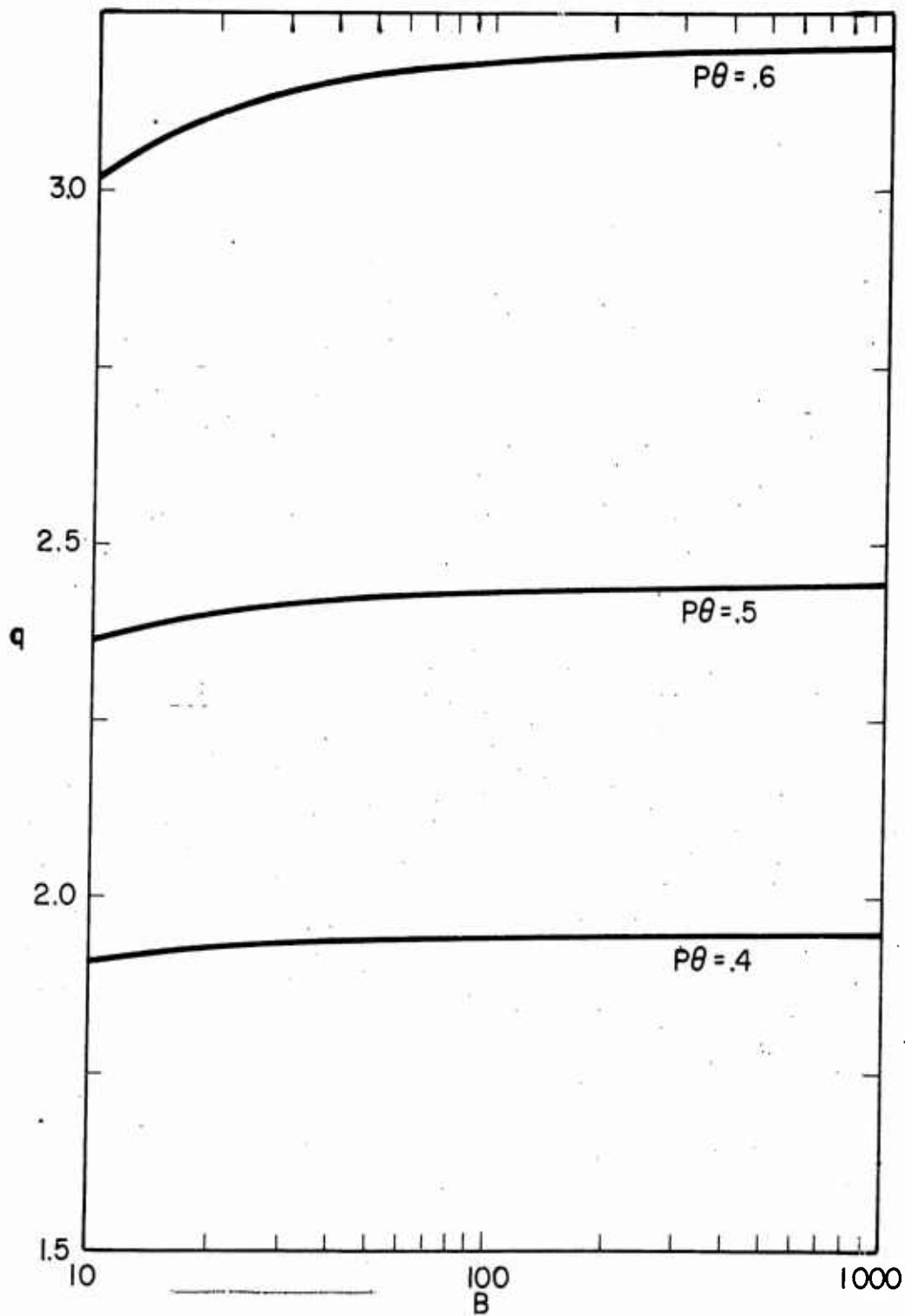


FIGURE 2. CRITICAL CASE; q OBTAINED BY MATCHING AT DIFFERENT VALUES OF $P\theta$

matching at $\theta = 1$, the midpoint. The resulting values of q for several values of A/δ are shown in Fig. 3 which plots q versus B/q . It should be noted that the curves end abruptly. Corresponding to (13), the relation

$$\frac{q}{B} = \sqrt{4.344 \frac{A}{\delta} - \left(\frac{A}{\delta} + .718\right)} \quad (15)$$

can be obtained, and for a given A/δ there are no supercritical points for values of B/q smaller than those implied by (15).

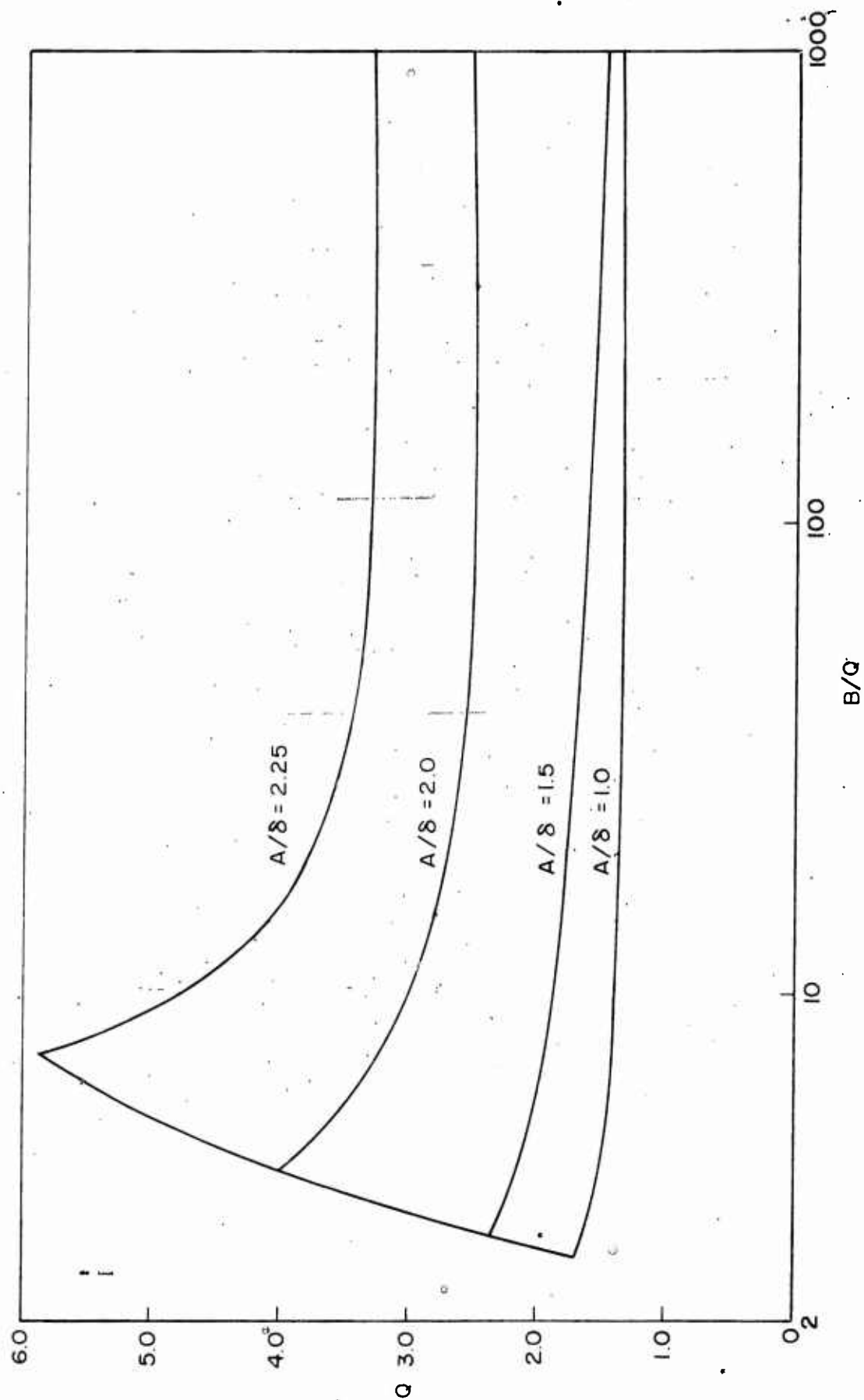


FIGURE 3. VALUE OF Q FOR VARIOUS A/δ OBTAINED BY MATCHING AT $\theta=1$ (SUPERCRITICAL CASES)

SPECIFIC RESULTS

The integral equation (6) was solved numerically for a number of sets of A/δ and B using $\Delta\theta = 0.1$. Some check calculations with smaller values of $\Delta\theta$ (0.05 and 0.01) indicated that the step size was sufficiently small. The results of these numerical calculations will be compared with the approximate analytical calculations of the critical conditions and ignition delay obtained in the previous section and those reported by Thomas³. Also, a comparison with experimental temperature histories obtained by Akita⁵ will be presented.

Critical Conditions

Fig. 4 compares the value of $\frac{\delta_{cr}(B)}{\delta_{cr}(\infty)} = e \div \left(\frac{A}{\delta}\right)_{cr}$ obtained from (13), the value of q being obtained by matching at $P\theta = 0.5$, with Thomas' expression³

$$\frac{\delta_{cr}(B)}{\delta_{cr}(\infty)} = 1 + 2.85B^{-2/3} \quad (16a)$$

and with Frank-Kamenetskii's expression⁶

$$\frac{\delta_{cr}(B)}{\delta_{cr}(\infty)} = 1 + 2.85B^{-2/3} \quad (16b)$$

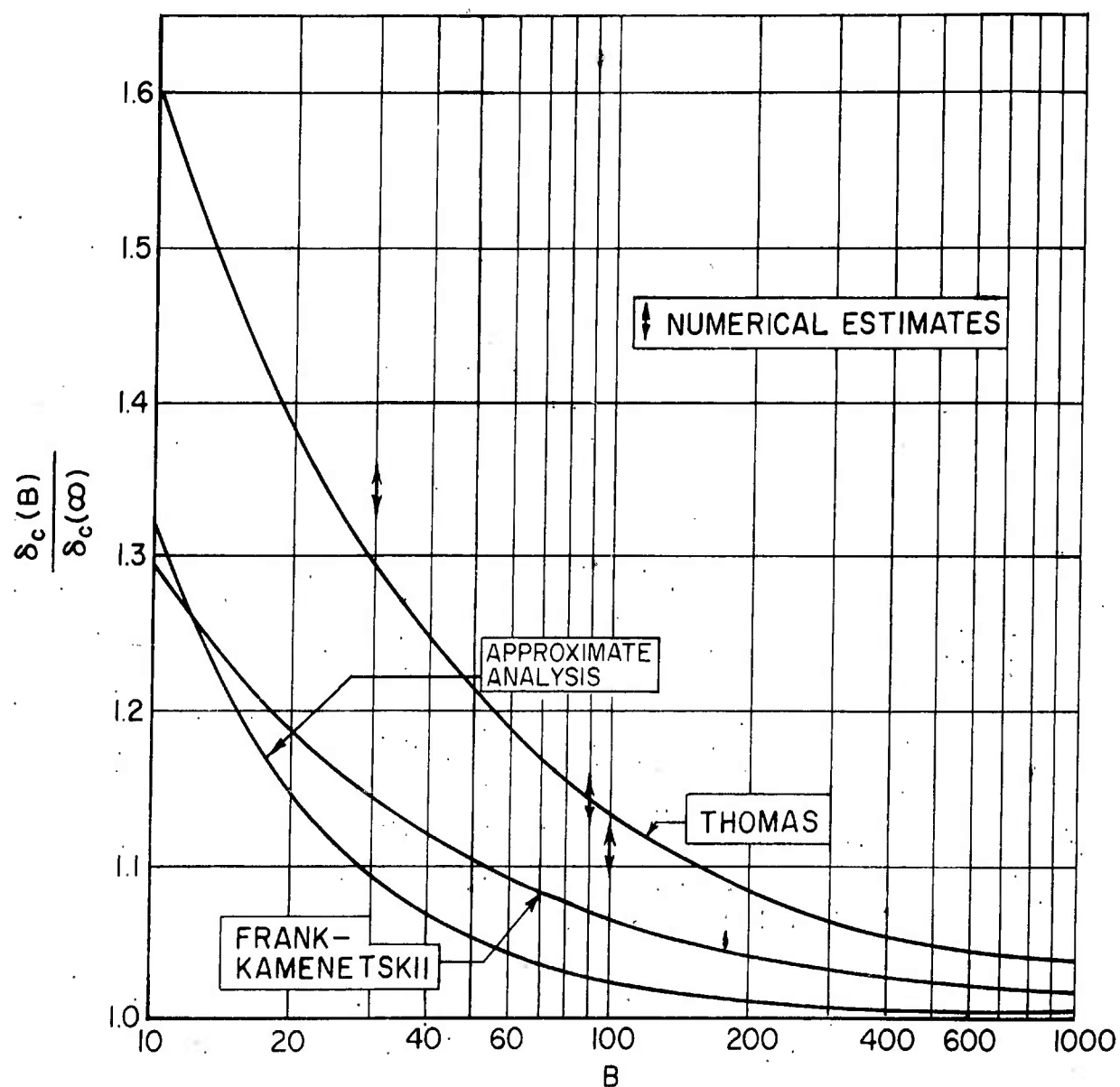


FIGURE 4. EFFECT OF REACTANT CONSUMPTION ON CRITICAL CONDITIONS FOR A THERMAL EXPLOSION

The limits of the stability curve found by the numerical calculations for particular values of B are indicated by \uparrow . While the limits were not determined closely except for $B = 176$, it is clear that Thomas' result is definitely superior to the others except for large B when the effect of reactant consumption is small.

Ignition Delay

The ignition delay τ_{ig} is defined somewhat arbitrarily as the time required for θ to reach 2. Fig. 5 shows $\delta\tau_{ig}$ as a function of A/δ for $B = 29.4$ and $B = 176$ as obtained by

- (1) Numerical solution of (6) combined with numerical evaluation of (4)
- (2) The approximate analysis based on (12b) with q determined by matching at $\theta = 1$
- (3) Thomas' approximate analysis, and
- (4) Numerical integrations carried out by Rice, Allen and Campbell⁷ in which, however, reactant consumption is not taken into account rigorously.

It can be seen that in this case our approximate analysis is in good agreement with our numerical calculations. The approximate analysis of Thomas and the numerical calculations of Rice, Allen and Campbell are somewhat further off, but in the opposite direction, than our approximate treatment.

Comparison with Experimental Temperature Histories

Kinbara and Akita⁴ have reported two temperature histories for sawdust spheres (contained in wire gauze) and compared the data with

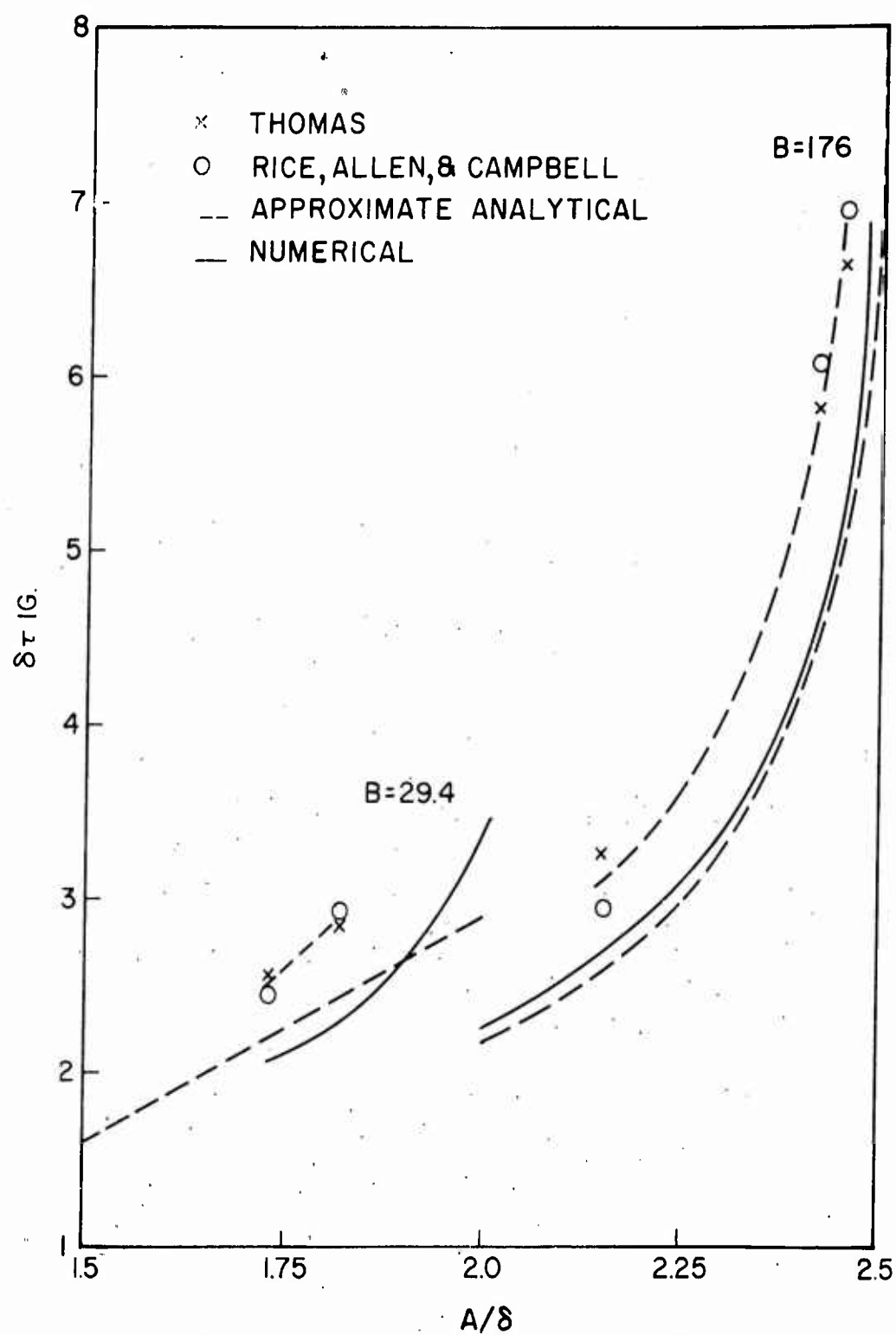
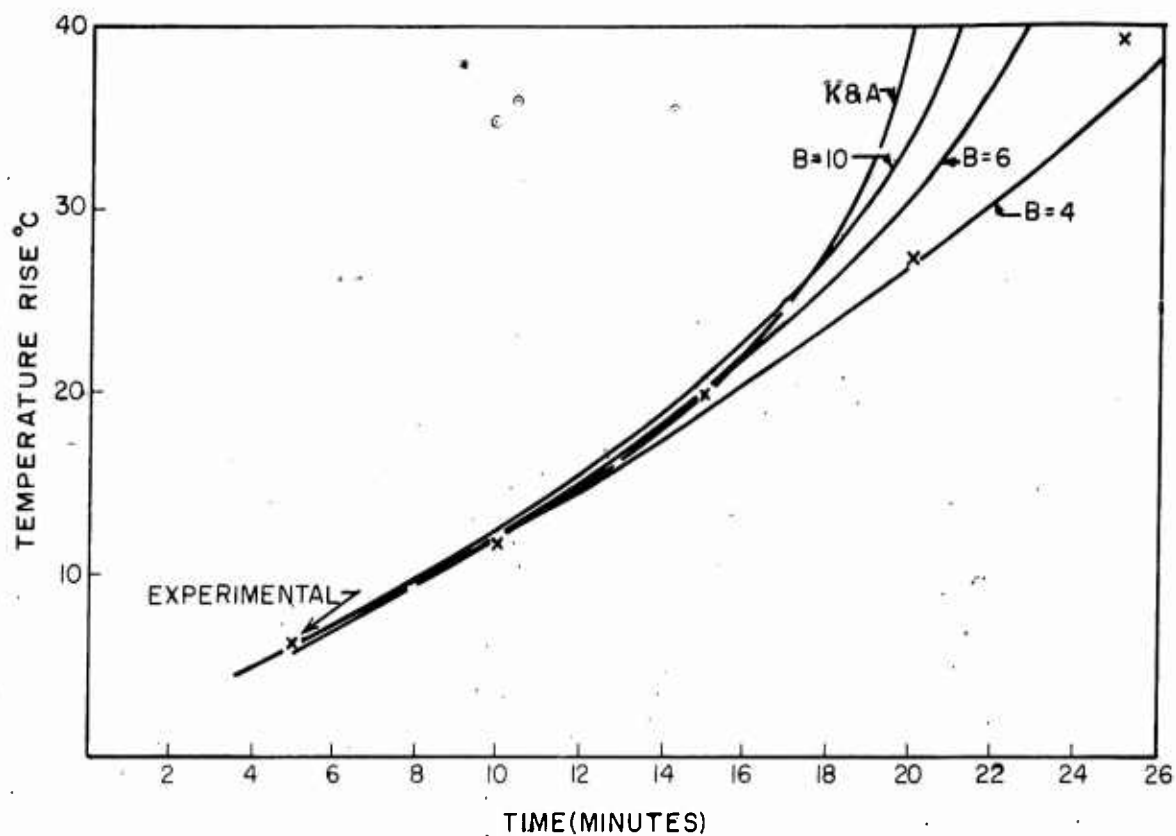


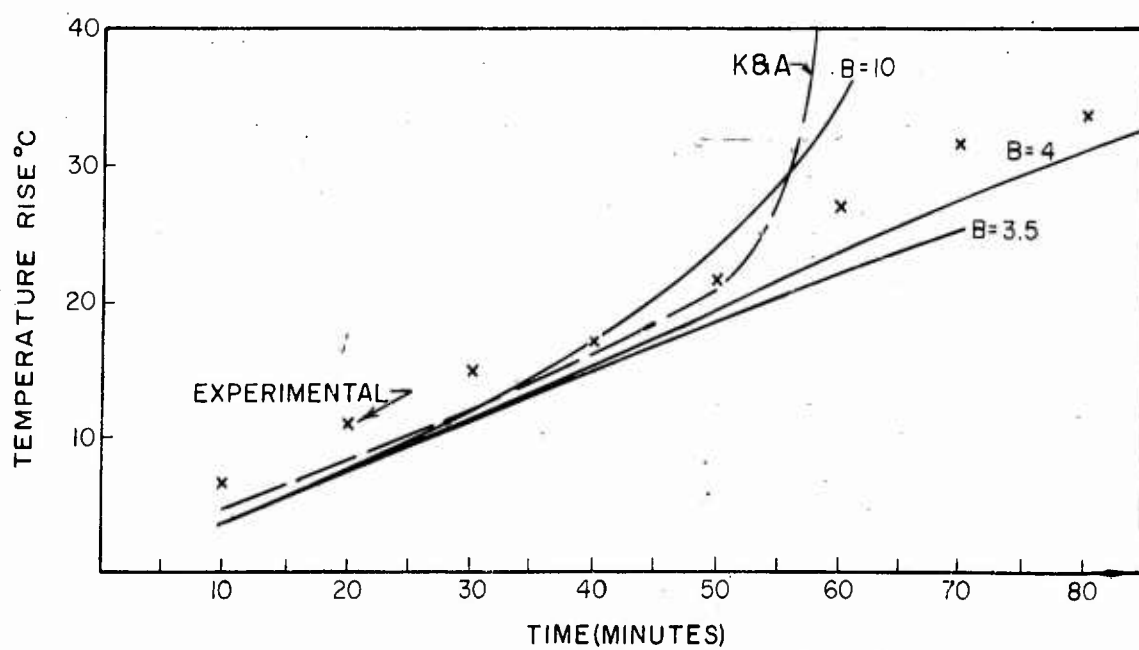
FIGURE 5. COMPARISON OF IGNITION DELAY RESULTS

solutions of a linearization of (1a). The agreement was qualitatively satisfactory, but the ignition delay was underestimated somewhat. It was suggested that this was due to the neglect of reactant consumption.

In Fig. 6a and 6b, the experimental results are shown along with Kinbara and Akita's calculated curves and numerical solutions for $B = 10$ and two lower values of B . It can be seen that a value of B of about 5 leads to a marked improvement in the agreement between theory and experiments. Unfortunately, Kinbara and Akita do not give the value of Q (the heat of decomposition of their sawdust), but $B = 5$ does correspond to a reasonable value.



6a—3.7cm SPHERE AT 196°C; $A/\delta = .46$



6b—5.25 cm SPHERE AT 179°C; $A/\delta = 0.65$

FIGURE 6. COMPARISON OF EXPERIMENTAL AND THEORETICAL TEMPERATURE HISTORIES

DISCUSSION

The mathematical analysis of the differential equations of non-steady explosion theory has been greatly simplified by using the temperature instead of the time as the independent variable. In particular, for a first order reaction, the numerical solution is greatly simplified because a nonlinear relation reduces to a quadratic which can be solved explicitly.

In the approximate analytical treatment of the equations, two approximations are involved. It is believed that the principal source of error is the use of the linear relation, (8), between composition and temperature as a first approximation to the solution rather than the use of the rational fraction, (11), to approximate the exponential. In particular, it could be anticipated that the linear approximation between composition and temperature would not be accurate near a temperature maximum. This probably explains the error in the determination of the critical conditions where a temperature maximum is involved and the relative accuracy of the determination of ignition delay for supercritical conditions where no maximum is involved.

It is, of course, true that, since there is an adjustable parameter, q , involved in the calculation, the determination of the critical condition could probably be improved by some other method of evaluating q . The method used in this paper of matching at a fixed value of $P\theta$ gave a value

of q which was essentially independent of B . An examination of Thomas' expression indicated that, to improve our result, q should increase as $B^{1/3}$, so that no basic improvement can be expected from changing the value of $P\theta$. While other methods of evaluating q are possible (such as matching at a fixed value of θ rather than $P\theta$), it hardly seems worthwhile to attempt to patch up the analysis in this way, particularly since the exact numerical solution is so convenient.

The comparison between Akita's experiments and the calculations indicate that the agreement is improved by taking reactant consumption into account, but, as Akita did not specify the value of Q for the sawdust, a question remains as to whether the value of B which gives satisfactory agreement is the actual value for the material though it appears reasonable. In view of the uncertainty in the measurements and in the value of the other properties involved, the agreement appears as good as can be expected. It would, however, be of some interest to treat the problem by numerical solution of the full partial differential equations (1a) and (1b) in order to estimate the effect of the approximations involved in nonsteady explosion theory. A numerical procedure for solving the partial differential equation has been developed⁸ but only a few preliminary calculations have been made as yet.

The numerical solutions for small values of B (≤ 10) show that the transition between the super- and subcritical cases becomes less

distinct as B becomes small. Examination of (14) shows that the maximum temperature in the subcritical case increases as B decreases, while the maximum temperature in supercritical cases must be less than B , so that the distinction obviously becomes less sharp for small values of B . In this region, it is necessary to use the numerical procedure rather than the analytical approximations which are essentially expansions in B^{-1} .

ACKNOWLEDGEMENT

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